Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713926090

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To cite this Article Schiller, P. , Pelzl, G. and Demus, D.(1987) 'Bistability and domain wall motion in smectic C phases induced by strong electric fields', Liquid Crystals, 2: 1, 21 — 30
To link to this Article: DOI: 10.1080/02678298708086634
URL: http://dx.doi.org/10.1080/02678298708086634

LIQUID CRYSTALS, 1987, VOL. 2, NO. 1, 21–30

Bistability and domain wall motion in smectic C phases induced by strong electric fields

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(*Received 10 March 1986; accepted 1 September 1986*)

The director configuration in smectic C phases is bistable in strong electric fields for suitable field directions with respect to the smectic layer planes. Therefore different domains separated by walls are possible. Bistability can be accompanied by director reorientation which causes domain wall motion and pronounced hysteresis effects. Transitions of this type have been investigated both experimentally and theoretically.

1. Introduction

As reported earlier [1–3], planar oriented smectic C liquid crystals with positive dielectric anisotropy sandwiched between parallel plates exhibit a Freedericksz transition which is characterized by a rotation of the director around the normal of the smectic layers. This transition occurs at a sharp electric threshold field, E_0, and no significant hysteresis is found. When the electric field is increased to very large values an irreversible texture transition appears based on a rearrangement of the smectic layers.

For suitable alignments of the smectic C phase in the cell another type of field-induced transition is found at fields above the Freedericksz threshold but below that for irreversible texture changes. This transition, based on a director reorientation, is visible as motions of domain walls and is accompanied by pronounced bistability effects, i.e. hysteresis. The aim of this paper is to explain the observed bistability above E_0. In the first section we propose a simple model describing bistabilities in thick samples of smectic C phases. Experimental results will be discussed in the final section.

2. Structure of smectic C phases

In smectic C phases molecules are arranged in a layered structure. Figure 1 shows the orientation of the director, $\mathbf{U}$, with respect to the smectic layer plane which is chosen to be identical to the xy plane of a cartesian system of coordinates. $\mathbf{U}$ can rotate around the layer normal (i.e. the z axis) without causing distortions of the smectic plane. On rotation, the angle θ between $\mathbf{U}$ and the layer normal remains unchanged so that $\mathbf{U}$ lies on a cone with aperture 2θ. The projection of $\mathbf{U}$ on the xy plane yields a vector $\mathbf{n}$. The rotation angle Φ varying between zero and 2π is defined by $\mathbf{n}$ and the x axis. For a ferro-electric smectic C phase a vector of spontaneous polarization $\mathbf{P}$ lies in the xy plane perpendicular to $\mathbf{n}$. It is convenient to introduce additionally an angle η enclosed by the x axis and $\mathbf{P}$.

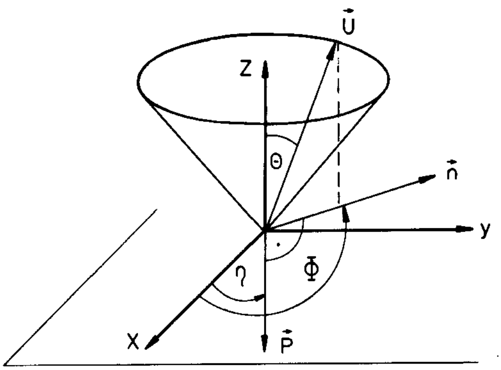


Figure 1. Orientation of the director with respect to the smectic plane (i.e. the xy plane). Here **U** is the director, θ is the tilt of the director on the layer normal, **n** is a unit vector oriented parallel to the projection of **U** on the layer plane, Φ is the rotation angle of **n**, and **P** is the vector of the spontaneous polarization, η is the rotation angle of **P**.

3. Domains in thick cells

Bistability effects have been found experimentally above the Freedericksz threshold E_0. Let us refer the theoretical description to the case of high electric fields $E \gg E_0$. At strong fields the plates on the sample surface only affect the director configuration within a thin boundary layer of thickness ξ which is much smaller than the cell thickness D

$$D \gg \xi. \tag{1}$$

The coherence length ξ is estimated by the formula

$$\xi = \sqrt{\left(\frac{B}{|\Delta\varepsilon|}\right)} \frac{\pi}{\sin\theta E}, \tag{2}$$

where B is an effective curvature elastic constant and $\Delta\varepsilon$ the difference of dielectric constants measured parallel and perpendicular to **U**. The inequality (1) defines the condition for a thick cell. In a theoretical treatment of bistabilities, thick cells can be regarded approximately as infinite media.

If bistability appears at least two domain types with different director orientations are present simultaneously. The domains are separated by walls with thickness comparable to the coherence length ξ. Note that there is also possible the occurrence of 2π walls between domains with the same director alignment. However we restrict our attention to walls separating differently oriented domains. Because walls may be oriented arbitrarily, we introduce an angle β between the normal **W** of a domain wall and the z axis (cf. figure 2). According to figure 2 the vector **E** of the electric field is assumed to lie in the xz plane tilted by an angle α with respect to the z axis. It should be emphasized that both α and β are polar angles with possible values in the range $0 \leqslant \alpha \leqslant \pi$ and $0 \leqslant \beta \leqslant \pi$, respectively. An azimuthal angle of **W** which is enclosed between the projection of **W** onto the xy plane and the x axis can also be defined. However, any results which we obtain do not depend on this angle.

As an example consider a planar aligned smectic C liquid crystal with positive dielectric anisotropy in a strong electric field ($E \gg E_0$). Except for thin boundary layers at the substrates the director is turned around the cone axis as far as possible in the field direction (cf. figure 3). In accord with the theoretical results of Rapini [4]

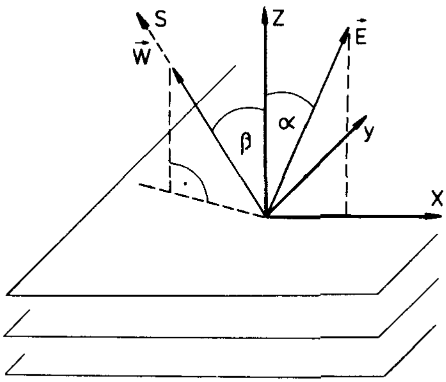


Figure 2. The angles α and β define the direction of the electric field **E** and the normal **W** of a domain wall, respectively. s is the axis of the coordinates oriented parallel to **W**.

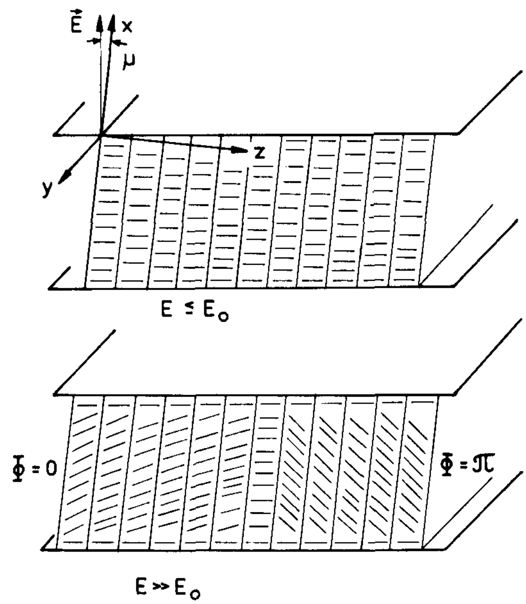


Figure 3. Planar oriented smectic C liquid crystal (μ is the tilt angle of the smectic layers with respect to the substrate normal). At $E \gg E_0$ two domain types with azimuthal angles $\Phi = 0$ and $\Phi = \pi$ are possible.

we assume that distortions of the smectic layer planes are negligible. Since the rotation of the director may be either clockwise or anticlockwise, two domain types are possible differing in the azimuthal angle Φ by π. Both states are separated by walls. If the smectic layer planes are tilted to the electric field by an angle μ (cf. figure 3), the free energies of the domains differ from each other. In this case walls between domains move with a constant velocity, which is calculated for the limit of strong electric fields in §5.

4. Free energy

The density of the curvature elastic free energy caused by director rotations contains four different elastic constants [4]. In a simplified model only two elastic constants are introduced which refer to distortions with gradients parallel and perpendicular to the smectic layers, respectively

$$f_1 = \tfrac{1}{2}B_{\parallel}\left[\left(\frac{\partial\Phi}{\partial x}\right)^2 + \left(\frac{\partial\Phi}{\partial y}\right)^2\right] + \tfrac{1}{2}B_{\perp}\left(\frac{\partial\Phi}{\partial z}\right)^2. \tag{3}$$

Applying equation (3) to a plane domain wall with normal **W** (cf. figure 2) we find

$$f_1 = \tfrac{1}{2}B\left(\frac{\partial\Phi}{\partial s}\right)^2, \tag{4}$$

where

$$B = B_{\parallel}\sin^2\beta + B_{\perp}\cos^2\beta$$

is an effective elastic constant. The density of the electric field energy is given by

$$f_2 = -\tfrac{1}{2}\Delta\varepsilon(\mathbf{E}\cdot\mathbf{U})^2. \tag{5}$$

Using angles defined in figure 2 the expression

$$f_2 = -\tfrac{1}{2}\Delta\varepsilon E^2(\cos\alpha\cos\theta + \sin\alpha\sin\theta\cos\Phi)^2 \tag{6}$$

is obtained. For a ferro-electric smectic C phase a second electric field term results due to the spontaneous polarization

$$f_3 = -\mathbf{P}\cdot\mathbf{E} \tag{7}$$

or, written explicitly for our special geometry

$$f_3 = -PE\sin\alpha\cos\eta. \tag{8}$$

The overall free energy

$$f = \int dV(f_1 + f_2 + f_3) \tag{9}$$

is a minimum when the system is in an equilibrium configuration; here V denotes the volume of the system.

5. Moving and fixed walls in a non-chiral smectic C phase

Let us regard two domains in the smectic C phase separated by a plane wall which are subjected to an electric field. The free energy per wall area is given by

$$F = \frac{1}{2}\int ds\left[B\left(\frac{\partial\Phi}{\partial s}\right)^2 + 2f_2\right]. \tag{10}$$

Beyond the walls Φ does not depend on s, and only f_2 determines the free energy. The stability of the domains requires that f_2 as a function of Φ possesses two minima corresponding to different director orientations. Analysing formula (6), bistability is found, when the inequality

$$\left|\frac{\cos\alpha\cos\theta}{\sin\alpha\sin\theta}\right| < 1 \tag{11}$$

is satisfied. Otherwise the system is monostable, that means either $\Phi = 0$ or $\Phi = \pi$ is the unique stable director configuration. Note that equation (11) is equivalent to

$$\left|\frac{\pi}{2} - \alpha\right| < \theta, \tag{12}$$

and so two types of bistability, determined by the sign of $\Delta\varepsilon$, have to be taken into account.

5.1. *Positive dielectric anisotropy*

If $\Delta\varepsilon > 0$ the configurations

$$\left.\begin{aligned}\Phi_1 &= 0\\ \Phi_2 &= \pi\end{aligned}\right\} \quad \text{and} \tag{13}$$

correspond to minima of f_2 (metastable and absolutely stable states). Except for the case $\alpha = \pi/2$ absolutely stable domains should grow at the cost of the metastable domains by motion of walls. To describe this time dependent behaviour, viscous friction forces are taken into account in the torque balance equation

$$-\lambda \frac{\partial \Phi}{\partial t} = \frac{\delta F}{\delta \Phi}, \tag{14}$$

where λ is the rotational viscosity [5] and t is the time. Using equations (10) and (6) we find explicitly

$$-\lambda \frac{\partial \Phi}{\partial t} + B \frac{\partial^2 \Phi}{\partial s^2} = a \sin \Phi + b \sin \Phi \cos \Phi, \tag{15}$$

where the abbreviations

$$\left.\begin{aligned}a &= \Delta\varepsilon E^2 \sin\theta \cos\theta \sin\alpha \cos\alpha\\ b &= \Delta\varepsilon E^2 \sin^2\theta \sin^2\alpha\end{aligned}\right\} \quad \text{and} \tag{16}$$

are introduced. We note that the inequality in equation (11) takes the form

$$\left|\frac{a}{b}\right| < 1. \tag{17}$$

Equation (15) has a simple soliton-like solution describing a moving wall

$$\Phi(s, t) = 2 \arctan \exp \left[\sqrt{\left(\frac{b}{B}\right)} (s - vt) \right]. \tag{18}$$

The velocity of domain walls is then

$$v = \sqrt{\left(\frac{B}{b}\right)} \frac{a}{\lambda} \tag{19}$$

or

$$v = \frac{\sqrt{(B\Delta\varepsilon)} \cos\alpha \cos\theta E}{\lambda}. \tag{20}$$

5.2. *Negative dielectric anisotropy*

At $\Delta\varepsilon < 0$ domains with orientation

$$\left.\begin{aligned}\Phi_3 &= \arccos\left(\frac{a}{|b|}\right)\\ \Phi_4 &= -\Phi_3\end{aligned}\right\} \quad \text{and} \tag{21}$$

(or equivalently $\Phi_4 = 2\pi - \Phi_3$) are stable; without loss of generality we can assume $0 < \Phi_3 < \pi$. As both states in equation (21) correspond to the same value of the free energy, domain wall motion cannot be generated. In this case the torque balance equation is obtained as

$$B\frac{\partial^2\Phi}{\partial s^2} = a\sin\Phi - |b|\sin\Phi\cos\Phi. \tag{22}$$

Assuming $s = 0$ at the middle plane of a wall, two solutions of equation (22) are found. The first

$$\Phi(s) = 2\arctan\left[\tan\frac{\Phi_3}{2}\tan h\left(\frac{1}{2}\sin\Phi_3\sqrt{\left(\frac{|b|}{B}\right)}s\right)\right] \tag{23}$$

satisfies

$$\left.\begin{aligned}\Phi(s\to-\infty) &= -\Phi_3,\\ \Phi(0) &= 0,\\ \Phi(s\to+\infty) &= +\Phi_3.\end{aligned}\right\} \tag{24}$$

The other solution

$$\Phi(s) = 2\arctan\left[\frac{\tan(\Phi_3/2)}{\tan h(-\frac{1}{2}\sin\Phi_3\sqrt{(|b|/B)}s)}\right] \tag{25}$$

is accompanied by

$$\left.\begin{aligned}\Phi(s\to-\infty) &= \Phi_3,\\ \Phi(0) &= \pi\\ \Phi(s\to+\infty) &= 2\pi-\Phi_3.\end{aligned}\right\} \tag{26}$$

Generally, both wall types differ somewhat in thickness and elastic energy.

6. Response of a ferro-electric smectic C phase with negative di-electric anisotropy

Ferro-electric smectic C phases offer more possibilities to achieve bistability by applying d.c. electric fields. We restrict our attention to the special case $\Delta\varepsilon < 0$ and the electric field direction parallel to the smectic layers (parallel to the x axis in figure 2). To unwind the original helical arrangement of the director in ferro-electric smectic C phases and to avoid electrohydrodynamic instabilities, the d.c. field E_1 should be combined with an appropriate a.c. field E_2,

$$E = E_1 + \sqrt{2}E_2\sin\omega t. \tag{27}$$

The frequency ω is chosen to be high enough, so that the director does not follow the alternating field

$$\omega \gg \frac{|\Delta\varepsilon|\sin^2\theta E_2^2 + PE_2}{\lambda}. \tag{28}$$

In this case the time averaged field energy is determined by

$$f_2 + f_3 = \tfrac{1}{2}|c|\sin^2\eta - d\cos\eta, \tag{29}$$

where

$$\left.\begin{aligned}|c| &= |\Delta\varepsilon|\sin^2\theta(E_1^2+E_2^2)\\ d &= PE_1.\end{aligned}\right\} \tag{30}$$

and

Now the torque balance takes the form

$$-\lambda\frac{\partial\eta}{\partial t} + B\frac{\partial^2\eta}{\partial s^2} = d\sin\eta + |c|\sin\eta\cos\eta; \tag{31}$$

depending on the ratio $R = |d/c|$ two different reorientation mechanisms can occur.

6.1. *Domain wall motion*

Introducing a parameter

$$R = \left|\frac{d}{c}\right| = \frac{PE_1}{|\Delta\varepsilon|\sin^2\theta(E_1^2 + E_2^2)}, \tag{32}$$

bistability is found for

$$R < 1. \tag{33}$$

Two domain types are possible which differ in the orientation of **P** with respect to the x axis

and

$$\left.\begin{array}{l}\eta_1 = 0\\ \eta_2 = \pi\end{array}\right\}. \tag{34}$$

η_1 and η_2 correspond to the metastable and absolutely stable configurations, respectively. The velocity, v, of walls between domains is obtained by the solution of equation (31)

$$\eta(s, t) = 2\arctan\exp\left[\sqrt{\left(\frac{|c|}{B}\right)}(s - vt)\right], \tag{35}$$

with

$$v = \sqrt{\left(\frac{B}{|\Delta\varepsilon|\sin^2\theta(E_1^2 + E_2^2)}\right)}\frac{PE_1}{\lambda}. \tag{36}$$

In ferro-electric smectic C phases the direction of the wall velocity can be reversed easily by reversing the d.c. field, E_1, as a consequence of the linear coupling of E_1 to the director torque.

6.2. *Bulk reorientation*

If R obeys

$$R > 1 \tag{37}$$

the unique, stable alignment of the polarization is $\eta = 0$. But bulk switching in the entire sample is generated by reversing E_1 (i.e. antiparallel to the x axis). In this case the reorientation of **P** from $\eta = 0$ to $\eta = \pi$ is determined by

$$-\lambda\frac{\partial\eta}{\partial t} = -d\sin\eta + |c|\sin\eta\cos\eta. \tag{38}$$

Assuming that at $t = 0$ the field E_1 is reversed, the solution of equation (38) for $t \geqslant 0$ is

$$\frac{PE_1}{\lambda}t = \frac{R}{2(R-1)}\ln\left(\frac{1-\cos\eta(t)}{1-\cos\eta(0)}\right) - \frac{R}{2(R+1)}\ln\left(\frac{1+\cos\eta(t)}{1+\cos\eta(0)}\right) - \frac{R}{R^2-1}\ln\left(\frac{R-\cos\eta(t)}{R-\cos\eta(0)}\right). \tag{39}$$

$\eta(0)$ is not equal to zero exactly, small deviations from the perfect alignment of **P** at $t = 0$ are caused either by a small misalignment of the smectic C phase or by thermal fluctuations. According to equation (39) at the beginning of the reorientation the initial disturbance $\eta(0)$ grows exponentially

$$\eta(t) = \eta(0)\exp\left(\frac{t}{\tau}\right), \tag{40}$$

with time constant

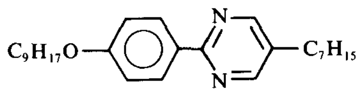
$$\tau = \frac{\lambda}{PE_1 - |\Delta\varepsilon|\sin^2\theta(E_1^2 + E_2^2)}. \tag{41}$$

The reorientation should be fast enough for electro-optic applications.

Experimental results concerning this process were published by Le Pesant *et al.* [6]. Switching times have been found in the range of a few milliseconds at very large fields.

7. Experimental observation of hysteresis in a non-chiral smectic C phase

The mesogen used in our investigations is 5-*n*-heptyl-2-[4-*n*-nonyloxyphenyl]-pyrimidine [7]



$T_{CS_C} = 45{\cdot}5°C$, $T_{S_CS_A} = 51°C$, $T_{S_AN} = 56{\cdot}5°C$ and $T_{NI} = 70°C$; this exhibits a positive dielectric anisotropy. An approximately planar director alignment was obtained by slowly cooling the well oriented nematic phase. Applying an electric field the Freedericksz transition occurs at a critical threshold, E_0. Above E_0 the director rotates around the layer normal, as far as possible in the field direction [2, 3]. In certain regions of the sample a kind of domain can arise which is metastable at high electric fields. In these domains above a second threshold E_0^* ($E_0^* > E_0$) a further director reorientation is performed gradually by domain wall motion. The velocity of the transition front clearly increases with increasing field strength. At sufficiently high fields ($E \simeq 6E_0$) the velocity is so large that moving walls can no longer be observed microscopically. The newly established high field state does not disappear when the electric field is gradually decreased below E_0^*. To switch off this state the field has to be decreased further below the Freedericksz threshold E_0.

Figure 4 shows the dependence of light intensity on the voltage curve for the original metastable domain and the new state which occurs above the second threshold E_0^*. In the smectic sample a dichroic dye is dissolved. The field-induced reorientation leads to an increase of light intensity in the wavelength region of the absorption (510 nm) of the dye. Obviously, the transition is accompanied by a pronounced hysteresis. Above 45 V the intensity curve is drawn by a dotted line, because at high fields the transition rate is so large that exact intensity measurements are not possible.

Figure 5 gives a geometrical interpretation of the observed bistability. The electric field **E** is always perpendicular to the substrate surface. Smectic layers which are parallel to the *xy* plane may be tilted by an angle μ with respect to **E**. According to the inequality in equation (12) the condition for bistability is

$$\mu = \left|\alpha - \frac{\pi}{2}\right| < \theta. \tag{42}$$

If $\mu < \theta$ both director configurations $\Phi = 0$ and $\Phi = \pi$ represent stable states at high electric fields. In figure 5 the state $\Phi = 0$ is metastable and $\Phi = \pi$ is absolutely stable. Small deviations from the planar alignment at the substrate surface can occur. When **U** is somewhat tilted out of the substrate plane as shown in figure 5, first the configuration $\Phi = 0$ appears at a fairly sharp Freedericksz threshold E_0. Increasing

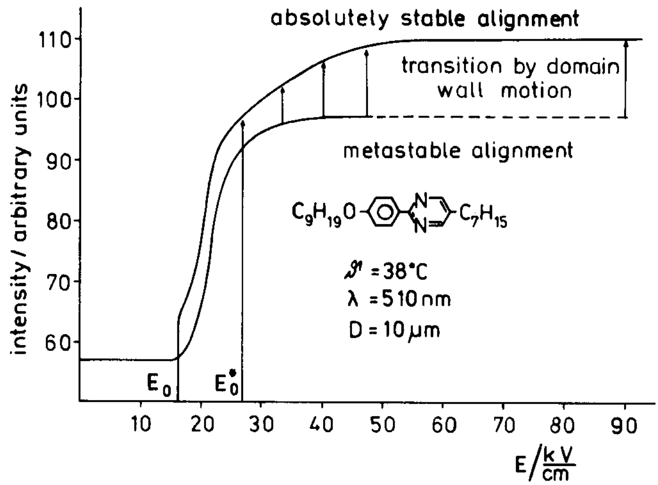


Figure 4. Light intensity versus voltage curves for the stable and metastable domains in the smectic C phase of 5-*n*-heptyl-2-[4-*n*-nonyloxyphenyl]-pyrimidine (ϑ = 38°C; wavelength 510 nm; D = 10 μm).

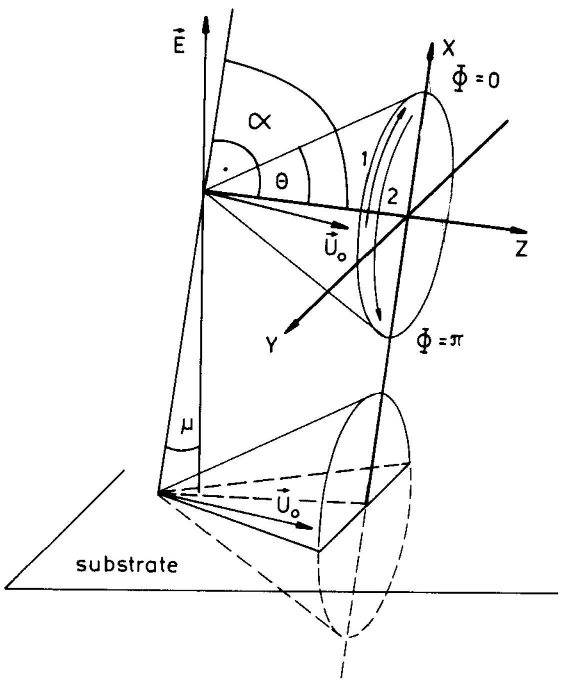


Figure 5. Switching processes: $\mathbf{U}_0$ shows the director in its initial position ($E < E_0$), there is a small deviation from an exactly planar alignment, μ is the angle enclosed by $\mathbf{E}$ and the smectic layers, the other angles and coordinate axes are defined as in figures 1 and 2; 1, switching of the director just above the Freedericksz threshold E_0; 2, second reorientation at high electric fields.

E further the director rotates to the state $\Phi = \pi$ which corresponds to the lowest electric field energy. This second transition is performed by domain wall motion as found theoretically (cf. §5.1).

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